Optical constants of ferromagnetic iron at the 2p resonances

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The energy dependence of optical constants at resonance represents a priviledged standpoint for comparing theory and experiment in terms of structural, electronic and magnetic properties of materials. In the X-ray range, this comparison is usually performed only over the imaginary part β of the complex refractive index $n = 1 - \delta - i\,\beta$, i.e. over the absorption. For decades, extended or near edge X-ray absorption spectra served as a reference for refining parametrized models of the local structure and of chemical environment. Over the last ten years the use of polarized x-rays has widened this field to include the study of anisotropic systems, and in particular of magnetic materials [1]. Model calculations, on the other hand, may deal with both the real and imaginary parts of the index at resonance. The real part is seldom exploited, due to a lack of experimental data, even if it contains as much information as the latter. It has even been shown that the interference between the two (as in resonant reflectivity) can facilitate the investigation of spectroscopy features, like satellites, that are weak but of major importance for understanding some ground state properties [2]. X-ray magneto-optics effects, related to the off-diagonal terms of the dielectric tensor, are stronger when the core excitations produce transitions to a final state that directly involves the "magnetic" orbitals, e.g. 3d for the transition metals of the first row or 4f for rare-earths. The largest cross-sections are observed for dipolar transitions, which implies that the most interesting resonances for studying magneto-optics effects (2p,3p -> 3d or 3d,4d -> 4f) are all located in the soft x-ray range. Most of the few direct experimental determinations of δ at resonance (i.e. without using Kramers-Kronig relations) have been performed on crystals by measuring the Bragg peak displacement when the photon energy is scanned through an absorption edge. The Bragg law imposes the relation between the photon wavelength λ and the crystal spacing 2d, hence, in order to match the 2d values for typical crystals (a few A), x-ray resonant magnetic scattering (XRMS) has been performed mainly at high energy. Recently, XRMS experiments have been performed with soft x-rays [3] using metallic multilayers as artificial periodic structures of adequate 2d spacing. We have determined the real part δ of the index of Fe across its 2p resonances by analysing the Bragg diffraction from an epitaxially grown Fe/V superlattice (2d = 30.6 A) prepared by sputter deposition of the metallic layers on an MgO (001) crystal [4]. We have compared our results to the only previous direct determination of δ obtained by measuring the Faraday rotation of x-rays transmitted by ferromagnetic iron [5]. X-ray scattering and absorption measurements were performed on the reflectometer of the soft x-ray metrology beamline 6.3.2 at ALS (Berkeley) [6]. We used the radiation emitted by a bending magnet between 110 and 200 μrad above the orbit plane, which gives elliptically polarized photons with a circular polarization rate of 40 % over the energy range 650-780 eV. The sample was magnetized parallel to its surface and in the scattering plane by means of a permanent magnet located behind the sample holder and mounted on a stepper motor used to reverse the field direction. Absorption spectra, recorded by measuring the photocurrent on the sample, were scaled to the non resonant values at 650 and 780 eV to give the imaginary part of the index $\beta^{\pm}$, where + and - signs refer to opposite magnetization/helicity orientations. Resonant scattering spectra are reported in Fig.1(a,b) as a function of θ and photon energy, for the two magnetization/helicity orientations. These are raw data, simply normalized to the incoming photon flux. Fig.1c shows the difference between the curves in a) and b). The decrement to the real part of the index, $\delta^{\pm}$, is obtained from the Bragg peak displacement versus energy following the model of Rosenbluth and Lee [7]. The four real functions ($\beta^{\pm}$ and $\delta^{\pm}$) allow to determine the helicity dependent refractive index $n^{\pm}$ for ferromagnetic iron at the 2p resonances in a strictly experimental way. The diagonal elements of the dielectric tensor are then given by $1/2[\,(n^{+})^2 + (n^{-})^2\,]$, and the off diagonal elements by $\pm\, i/2[(n^{+})^2 - (n^{-})^2\,]$. We consider that the experimental determination of the dielectric tensor including its off-diagonal elements is an important step towards a better refinement of theoretical and calculational models that describe the electronic and magnetic ground state properties of materials. To this end, it is essential to develop several independent experimental methods in addition to the standard Kramers-Kronig approach. Polarization rotation (upon transmission or reflection), interferometry and resonant magnetic scattering together constitute a sound basis for future work.

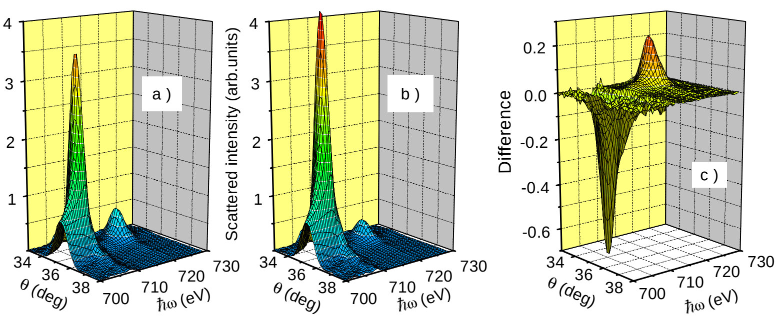

Fig.1. θ/2θ scans at various photon energies (2p edges of Fe) for parallel (a) and antiparallel (b) magnetization/helicity orientations. Panel (c) reports the corresponding difference curves.

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